Electron tunneling time measured by photoluminescence excitation correlation spectroscopy

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The tunneling time for electrons to escape from the lowest quasibound state in the quantum wells of GaAs/AlAs/GaAs/AlAs/GaAs double-barrier heterostructures with barriers between 16 and 62 Å has been measured at 80 K using photoluminescence excitation correlation spectroscopy. The decay time for samples with barrier thicknesses from 16 Å (≈ 12 ps) to 34 Å (≈ 800 ps) depends exponentially on barrier thickness, in good agreement with calculations of electron tunneling time derived from the energy width of the resonance. Electron and heavy hole carrier densities are observed to decay at the same rate, indicating a coupling between the two decay processes.

The electrical properties of the double-barrier (DB) heterostructure have been of great interest since its proposal by Tsu and Esaki.[1] The desire to characterize the high-frequency behavior of the DB stems from interest in its use as an oscillator[2] and as a switching element.[3,4] However, the time associated with the tunneling of electrons has been the subject of more than 20 years of discussion.[5–9] Experimental measurements of tunneling times have required the development of high-speed measurement techniques. Recently, two groups have reported experimental studies of the temporal response of DB structures. Whitaker *et al.*[10] have used electro-optic sampling measurements to study a single tunnel device. Tsuchiya *et al.*[11] used the photoluminescence (PL) from carriers in the quasibound states in the quantum well to study the decay of the electron population in the quantum well as a function of the barrier thickness. However, the experiments of Ref. 11 were limited to times greater than 60 ps, and Guo *et al.*[9] have raised some serious questions about the theoretical calculations that were used for comparison with experimental results.

In this letter we report a study of the decay of photoexcited carriers in double-barrier heterostructures as a function of the thickness of the barrier layers. By employing an excitation correlation method[12] we have extended the results of Tsuchiya *et al.*[11] to significantly shorter times and are comparing our data with results of corrected calculations of the tunneling times.

Double-barrier (DB) structures were grown on (100) GaAs substrates by molecular beam epitaxy in a Perkin–Elmer 430 system at 600 °C. After growth of 0.5 μm of GaAs, a superlattice buffer layer consisting of five periods of 50 Å $Al_{0.35}Ga_{0.65}As$/500 Å GaAs was grown. This was followed by growth of a 0.7 μm layer of GaAs, which provided a high quality layer on which to grow the DB and eliminated any optical effects from the superlattice. Then a symmetrical GaAs/AlAs/GaAs/AlAs/GaAs DB was grown, with a well thickness of 58 Å. The final layer was a 300 Å GaAs cap. All layers were nominally undoped with an estimated residual carbon acceptor concentration of 10^{14} cm^{-3}. Seven samples were studied, with bulk growth rate information predicting barrier thicknesses of 16, 22, 28, 34, 34, 48, and 62 Å. High-resolution transmission electron microscopy confirmed the barrier thicknesses of the 16 Å sample and one of the 34 Å samples, within an uncertainty of two monolayers. We estimate an uncertainty in barrier thickness of two monolayers for all of the samples.

The experimental technique has been described previously.[12] A colliding pulse mode-locked ring dye laser is used to generate a train of pulses 200 fs full width at half maximum (FWHM), at a repetition frequency of 120 MHz. The laser output is centered at 6200 Å and has a spectral width of 20 Å FWHM. The pulse train is equally divided into two separate beams which are independently chopped at $f_1 = 1600$ Hz and $f_2 = 2100$ Hz, and delayed with respect to one another by time γ ($-500 \leqslant \gamma \leqslant 500$ ps) before being recombined and focused to a 25-μm-diam spot on the surface of the sample. The typical average power used was 1 mW per beam before chopping. The PL is spectrally resolved, and detected by a GaAs photomultiplier tube (PMT). After amplification, the PMT signal is synchronously detected by a lock-in amplifier at either the fundamental frequency f_1 or at the sum frequency $f_{sum} = f_1 + f_2$. All of the results reported here were taken with the sample at 80 K.

In Fig. 1 we present a schematic diagram of the pro-

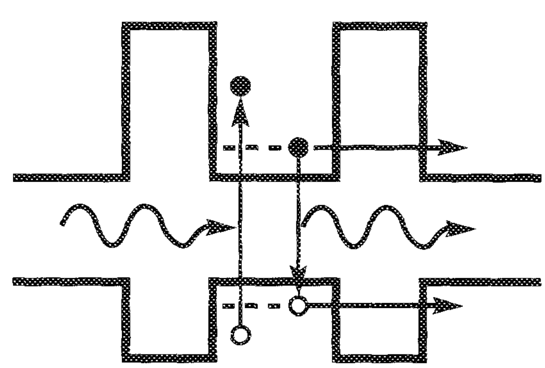

FIG. 1. Schematic diagram of relevant carrier processes involved in the double-barrier samples during the experiment. Shown are photoexcitation of carriers in the well, tunneling of carriers out of the well, and recombination of carriers in the well.

cesses of excitation, tunneling from the well, and the radiative recombination of carriers within the well. Recent observations of the thermalization times of electrons between subbands have given times less than 200 fs.[13] Hence we will assume that the thermalization of electrons and holes to the lowest subband to be fast compared to the times of interest here. A model of the measurement process has been given in Ref. 12. We will simply sketch the important results. If the electron and hole populations created by an optical pulse are $n(t)$ and $p(t)$, and the populations created by two optical pulses are independent, then the sum frequency signal $I_{\rm sum}$ is proportional to the cross correlation

$$I_{\rm sum}(\gamma) \propto \int [n(t)p(t-\gamma) + n(t-\gamma)p(t)]dt.$$

This signal is due to the recombination of electrons created by the first pulse with holes created by the second pulse, and vice versa. If we assume that the electron and heavy hole densities decay exponentially[7] with time constants τ_e and $\tau_{\rm hh}$, respectively, then the sum frequency signal is proportional to the sum of two exponentials

$$I_{\rm sum}(\gamma) \propto [\exp(-|\gamma|/\tau_e) + \exp(-|\gamma|/\tau_{\rm hh})]. \quad (1)$$

We have used this result as the basis for interpreting our data.

In Fig. 2 we present typical photoluminescence spectra taken at 80 K at the fundamental and sum chopping frequencies. The spectrum at the fundamental frequency consists of a single distinguishable feature centered at 7650 Å. The wavelength of the feature in the fundamental frequency spectrum is in approximate agreement with the calculated position of 7730 Å for transition from the lowest electron subband to the lowest heavy hole subband in a 58 Å quantum well. The peak of the corresponding feature in the sum frequency spectrum is shifted slightly to longer wavelengths. We do not, at present, have an explanation for this shift.

In Fig. 3 we present a semilogarithmic plot of a typical scan of the photoluminescence at the sum chopping frequency as a function of the time delay between the two pulses. The scan was taken at 80 K, at a wavelength of 7665 Å, the peak of the sum PL spectrum shown in Fig. 2. This delay scan consists of a peak at zero delay with wings extending to much longer times. The peak at $\gamma = 0$ is a coherence peak due to the optical interference of the two incident pulses on the sample and is not resolved in this scan. A single exponential fits the wings in the sum frequency delay scan shown in Fig. 3. Delay scans for one sample, taken at 4 K, show evidence of a second exponential with a faster decay. Fits to the sum frequency delay data at 80 K using a single exponential are shown as dashed lines in Fig. 3. The coherence peak is not included in fitting the delay scans.

In Fig. 4 we have plotted the exponential decay time at 80 K as a function of barrier thickness for the seven samples described above. The decay time depends exponentially on barrier thickness for barriers up to approximately 34 Å. Over this range of exponential dependence the decay time varies by two orders of magnitude. For thicker barriers the decay time seems to approach a value that is independent of the barrier thickness. With the 120 MHz repetition rate of the excitation pulses, there is an upper limit on the order of 2 ns to the decay times that can be measured accurately with our technique. Consequently, the result for the sample with a barrier thickness of 62 Å should be viewed with some cau-

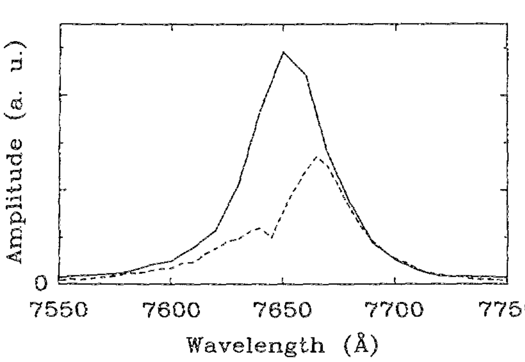

FIG. 2. Typical excitation correlated luminescence for 28 Å barrier sample at 80 K. Shown are luminescence signals at the fundamental chopping frequency (solid line) and the sum frequency (dashed line). Both scans were taken with delay $\gamma = 0$. The sum frequency spectrum has been multiplied by 2.

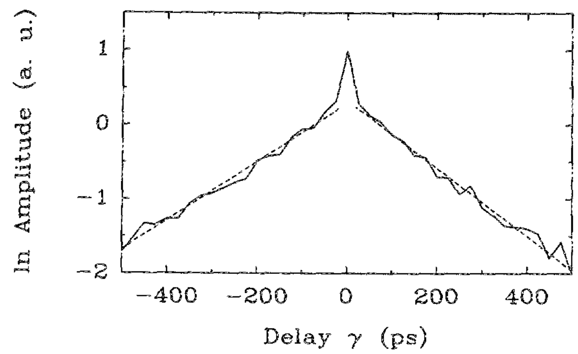

FIG. 3. Semilogarithmic plot of the variation of the sum frequency luminescence signal (solid line) with delay γ. The sample is the same as in Fig. 2. The scan was taken at 7665 Å, the peak of the sum frequency spectrum. The coherence peak at $\gamma = 0$ is not resolved in this scan. The dashed lines are the fits discussed in the text, which give a decay time of 236 ± 20 ps.

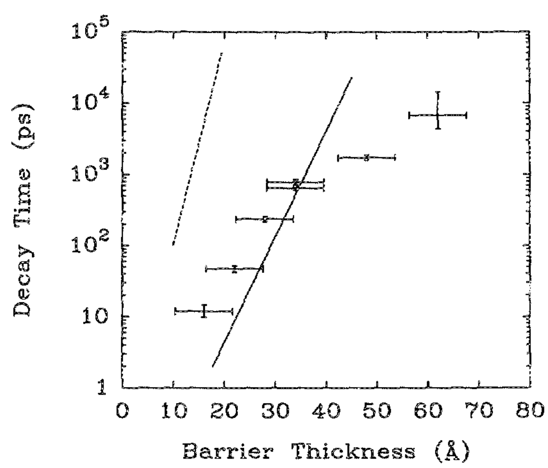

FIG. 4. Measured decay times as a function of barrier thickness. The data points are the measured decay times, with error bars on the thickness based on uncertainty in the barrier thickness and error bars on the decay times from uncertainties in the fits to the delay scans. The solid line is the electron tunneling time calculated from the width of the lowest conduction-band transmission resonance, using a two-band expression for the wave vector in AlAs barriers. The dashed line shows the tunneling time for heavy holes and was calculated using a one-band expression for the barrier wave vector.

tion. Our measured decay times are about a factor of 4 longer than those of Tsuchiya *et al.*[11]

In Fig. 4 we have also plotted the time calculated for electrons and heavy holes to tunnel out of the quantum well. The solid line is the electron tunneling time, and the dashed line is the heavy hole tunneling time. We will assume that the light holes thermalize to become heavy holes on a time scale short compared to times of interest here. From Ref. 7 the time for a particle to tunnel out of a quasibound state in the quantum well is related to the energy width of the corresponding resonance in the transmission probability by $\tau = \hbar/\Delta E_{\rm FWHM}$. The transmission probability is calculated using the transfer matrix approach of Kane,[14] modified to account for the different effective masses of the particle in the quantum well and in the barrier.

For electrons, we have considered only Γ-point barriers. It is appropriate to use a simple one-band expression for the wave vector in the well, $k = (2m_w^* m_e E/\hbar^2)^{1/2}$, where m_w^* is the effective mass in the GaAs well, m_e is the free-electron mass, and E is the energy of the particle with respect to the GaAs band edge. With the pure AlAs barriers in our samples, the lowest quasibound electron state has an energy far from the band edge in the AlAs barriers. Hence we have used a two-band model[15] to calculate the wave vector in the barriers. The barrier height used in these calculations was 1.07 eV, corresponding to a valence-band offset of 0.55 eV[16] and an AlAs band gap of 3.13 eV. The effective masses in the well and the barriers were taken to be $0.067m_e$ and $0.15m_e$, respectively.[17] For the heavy holes, we used a one-band expression to estimate the wave vector in the barriers. From the theoretical curves in Fig. 4, we can see that the tunneling time for electrons is much shorter than that for heavy holes.

Comparing these theoretical estimates of the tunneling times with the decay times observed experimentally, we note that the decay times agree well with the shorter tunneling time of the electrons. In contrast, if we expected Eq. (1) to explain the data, then we should observe two decay times, a short one near zero delay and a longer time at much longer delays. We might expect the longer decay time to be that for the heavy holes. There are a number of phenomena that could contribute to the observation of a single time that is close to the electron tunneling time. First, with the widely differing decay rates for electrons and heavy holes, it is very likely that the quantum well becomes charged. Rapidly escaping electrons could leave slowly decaying holes behind. The areal densities of holes and electrons produced in our experiment are roughly 10^{11} cm^{-2}, which are high compared to the background doping of 10^8 cm^{-2}. These areal densities produced by the photoexcitation could result in very substantial amounts of band bending. We would expect the potential across the barriers due to charging created by the differences in escape rates to increase the tunneling escape rate for holes. This effect would tend to make the electron and hole densities in the well follow each other and produce a decrease in the tunneling time for holes. Second, at higher temperatures, we would expect significant occupancy of the lowest light hole subband. The tunneling time for the light holes is even shorter than that for electrons and, hence, could offer an escape channel for the heavy holes. This phenomenon could be responsible for the changes in the observed delay scans with temperature. Finally, if we couple the finite spot size of the laser with the variations in decay rates, we might expect density gradients and electric fields to develop in the plane of the quantum wells. These density gradients and electric fields could act to make the hole density follow the electron density by inducing hole transport in the plane of the quantum well. We are attempting to carry out a detailed numerical simulation of this complex problem which will be important in understanding the results of any of the measurements reported to date that are based on optical excitation.

In summary, the tunneling time for electrons to escape from the lowest quasibound state in the quantum wells of GaAs/AlAs/GaAs/AlAs/GaAs double-barrier heterostructures has been measured at 80 K using the technique of photoluminescence excitation correlation spectroscopy. Results are in good agreement with calculations of electron tunneling time based on the energy width of the resonance in the transmission coefficient.

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